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Measurements of the gamma radiations from Br^{82} by the method of external conversion

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With 5 figures in the text

Introduction

During the last 15 years several investigations of the decay of Br^{82} have been reported by various authors (1–5) and some decay schemes have been proposed, the most recent being that of D. C. Lu *et al.* (6). The present paper deals with some measurements undertaken in order to obtain precise energy values and estimates of the relative intensities of the gamma-rays of Br^{82}. The experiments were performed in a large double-focusing beta-spectrometer of 50 cm mean radius.

Experimental

0.30 g NH_4Br was compressed into the shape of a cylindrical rod (4 × 10 mm) and was irradiated in a reactor at Harwell for 1 week to a maximum activity of about 100 mC. The Br^{82} source was then fixed in a brass cylinder which was carefully sealed to prevent gaseous bromine from escaping. For the calibration of the spectrometer a 300 mC Cs^{137} and a 20 mC Co^{60} source, were used. The material surrounding all activities was thick enough for a complete suppression of the primary beta-rays. For external conversion of the gamma radiations, a 3 mg/cm² uranium foil was used. This foil was prepared according to a recent method (7) of painting successive layers of an alcoholic solution of uranyl nitrate onto a thin aluminium backing and converting the nitrate by heating to uranium oxide. The thickness of the uranium layer was determined by weighing. By placing X-ray films in direct contact with the uranium, and exposing for about 20 hours, a blackening of the film was obtained, which showed that the homogeneity of the uranium converter was satisfactory. The size of the converter was 8 × 18 mm². The height (18 mm) was essentially taken to match that of the slit of the spectrometer GM-tube, while the width was chosen so as to allow electrons emitted at an angle of about 20° with respect to the direction of the incident gamma-ray to be accepted by the spectrometer aperture. This choice of angle was based on experimental determinations of the angular distribution of photoelectrons at different gamma-ray energies (8, 9) performed with the same spectrometer. The converter arrangement is shown schematically in Fig. 1.

The location of the sources in the spectrometer could be very accurately reproduced, as described by Hedgran (10).

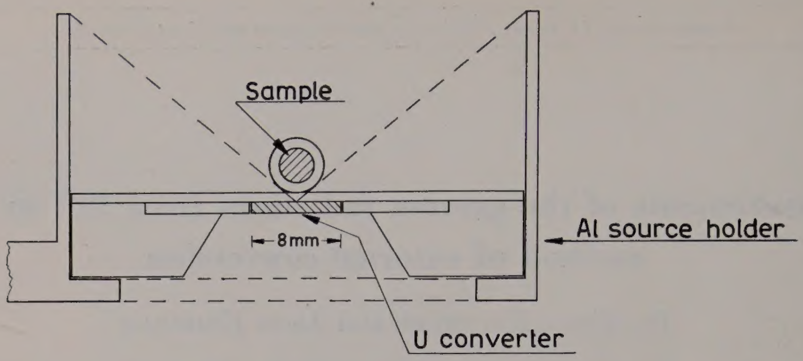


Fig. 1. Schematic view of source-holding arrangement.

Measurements

The external conversion lines from Br^{82} were measured in the double-focusing spectrometer with the arrangement outlined above. After some preliminary runs with lead and uranium foils of different thicknesses a 3 mg/cm^2 U foil was chosen as giving a good resolution and photo-lines of convenient intensities. The magnet current of the spectrometer was always varied in only one direction, which meant that the Br^{82} sample was taken out and the calibration sources were put in at their appropiate locations in the energy spectrum. Three runs were made.

Results

A typical set of photo-lines, obtained in one run, is shown in Fig. 2. These curves, together with the ionization chamber measurements, which were undertaken in order to determine experimentally relative photoelectric cross-sections, give precise values of the gamma-ray energies and information about the relative gamma-ray intensities.

All photo-lines were checked with respect to the known half-life of Br^{82} and were found to give values within 10 % of the common figure 35,7 hours (11). This value was then used for correcting the counting rates for decay.

a) *Precise determination of the gamma-ray energies.*—The actual gamma-ray energies range from 550 to 1475 keV. Thus, for calibration for the energy measurements, the 662 keV gamma-ray of Cs^{137} and the 1332 keV gamma-ray of Co^{60} were used as standards. Although the linearity of the spectrometer is known to be good, high accuracy requires more than one calibration energy, because the energy loss of the electrons in the converter cannot be neglected. The Co^{60} and Cs^{137} gamma-rays are sufficient for the purpose, since they lie near the extreme energy values of the region of interest. Roughly, the average electron energy loss in the range 0.3–3 MeV is constant at about 1 keV per mg/cm^2 in the case of U. More precisely, it decreases somewhat with increasing energy up to electron kinetic energies of about 1200 keV, according to Heitler (12). The effects of energy loss and scattering in the converter are reflected in the observed shift of the calibration factors, as shown in Fig. 3. Negligible amounts of these effects would have given a horizontal line in this energy region.

Table 1. Gamma-ray energies from the decay of Br^{82} as measured by external conversion in a 3 mg/cm^2 U foil.

Line	Energy in keV
Br^{82}: 1	554.1 ± 0.2
2	618.7 ± 0.2
3	698.4 ± 0.3
4	776.9 ± 0.2
5	827.6 ± 0.4
6	1044.0 ± 0.2
7	1317.1 ± 0.2
8	1475.3 ± 0.4
Standards:	
Cs^{137}	661.64
Co^{60}	1332.5

The deviation from constancy in the $H\varrho/\Omega$ value is thus not serious, and, because of the smallness of the effect, may be corrected for by linear interpolation between the calibration constants obtained for Cs^{137} and Co^{60}, as shown in Fig. 3. From this diagram the appropriate factor for conversion of Ω to $H\varrho$ was then obtained. The Ω value for every line was found by extrapolating the sides of the line and reading the abscissa of the intersection, because these parts of the conversion lines were generally best determined. The energies of the standard lines and the K-shell binding energy of U were taken from reference (13), pages 227 and 914. The energy values so obtained are shown in Table 1.

Table 1 is the result of three measurements. The errors have been estimated from the reproducibility of the values but it is assumed that they will also include possible systematic errors which will necessarily be small because of the calibration procedure adopted.

b) *Intensity measurements.*—According to Mitchell, reference (13), p. 236, the area A per unit momentum interval under a given photoelectron line can be written as

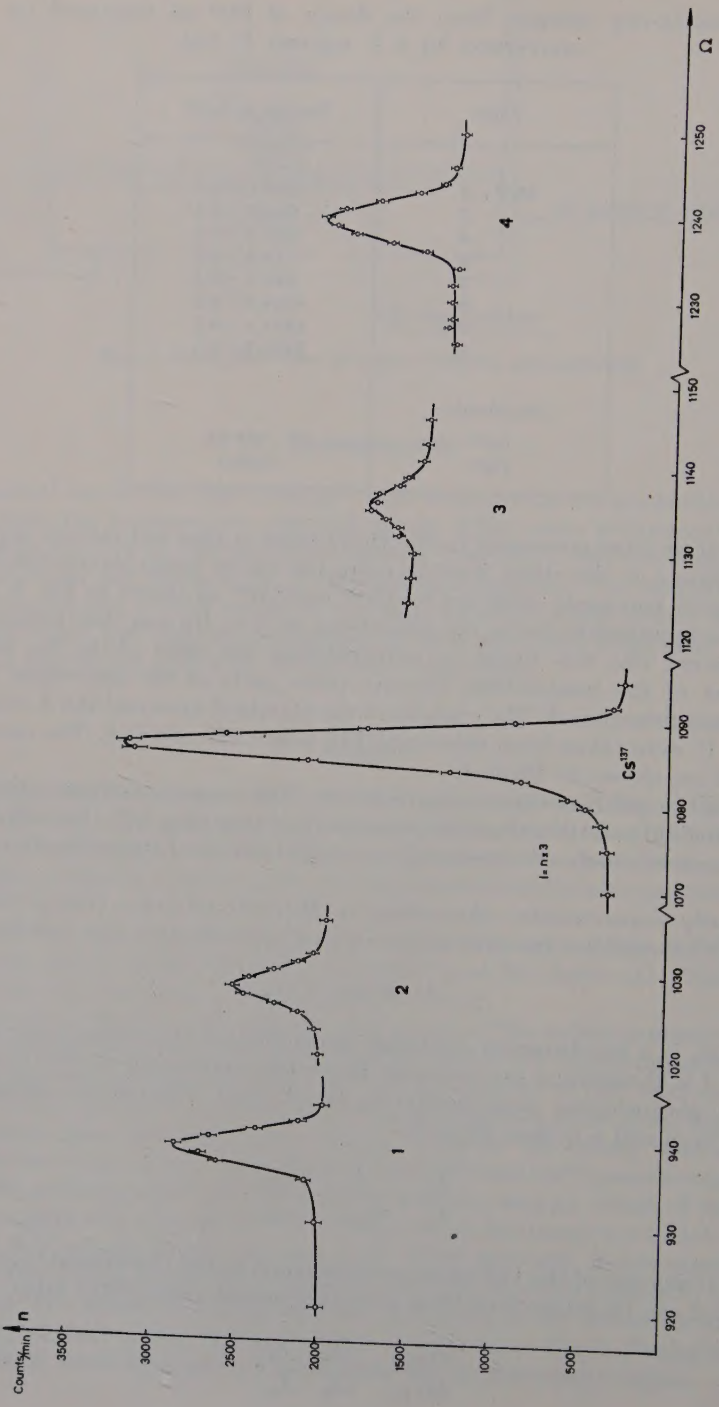
$$A = \text{const} \cdot I \cdot \tau_K, \tag{1}$$

where const. is a spectrometer constant, depending on the transmission and sample geometry, I is the gamma-ray intensity in arbitrary units and $\tau_K(E_\gamma)$ is the energy-dependent photoelectric cross-section for the K-shell. The relative intensity of two gamma-rays m and n is then given by

$$\frac{I_m}{I_n} = \frac{A_m}{(\tau_K)_m} \cdot \frac{(\tau_K)_n}{A_n}. \tag{2}$$

Thus, putting one of the two cross-sections equal to the theoretical value, the other cross-section can be determined from an experimental value of the ratio

$$\frac{(\tau_K)_m}{(\tau_K)_n} = \frac{A_m}{A_n} \cdot \frac{I_n}{I_m}. \tag{3}$$


Counts/min
n
500
1000
1500
2000
2500
3000
3500
920
930
940
1020
1030
1070
1080
1090
1120
1130
1140
1150
1230
1240
1250
Ω
1
2
I = n × 3
Cs137
3
4

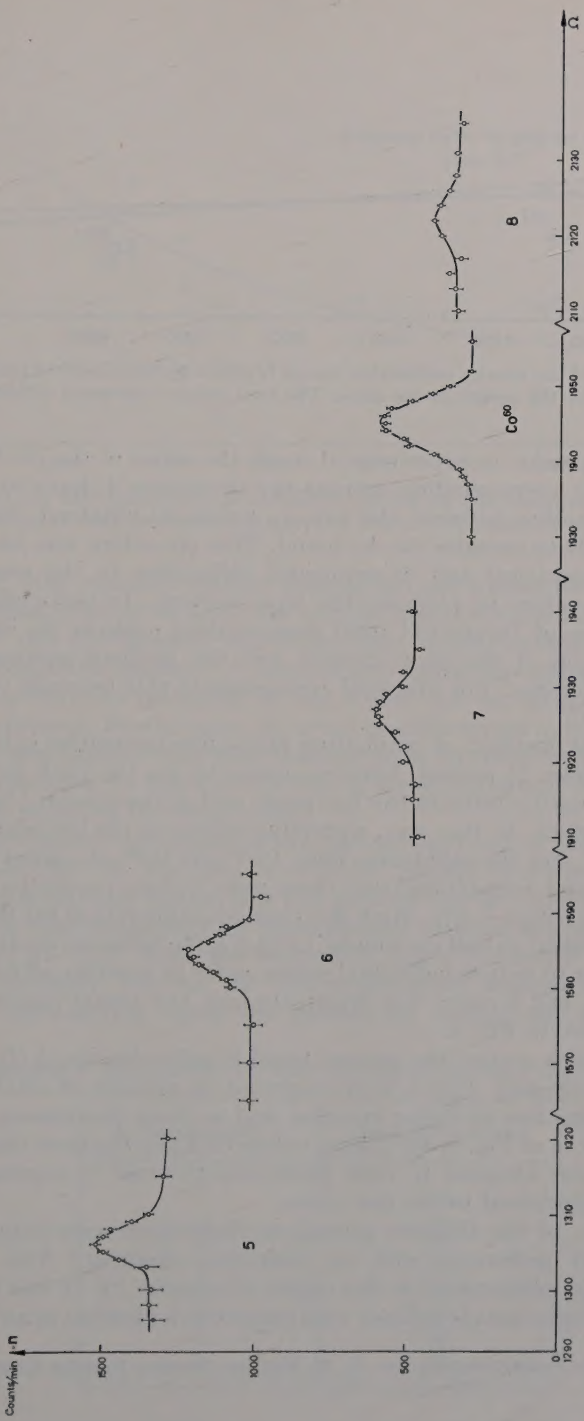


Fig. 2. External conversion lines from Br^{82}, Cs^{137} and Co^{60}, corrected for decay. 3 mg/cm^2 U converter.

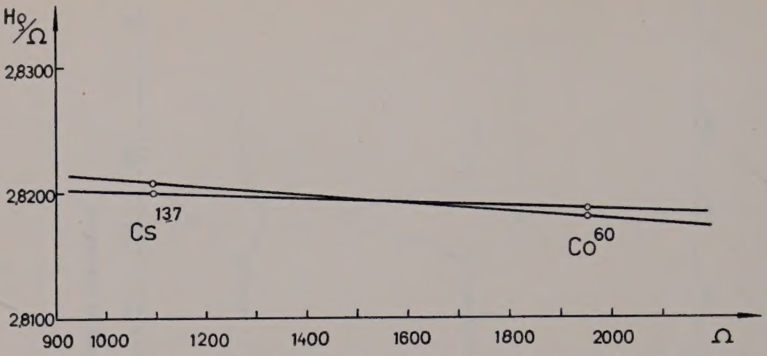


Fig. 3. The variation of the energy calibration factor $H\varrho/\Omega$ with the observed instrumental quantity "ohms", for the peaks of the lines. The two curves represent different runs.

Consequently, to make an experimental check the ratios of the photo-line intensities A, and of the corresponding gamma-ray intensities I, have to be found. By interpolating a function between the two τ_K values so obtained, "semiempirical" values for intermediate energies can be found. This procedure was adopted because of the inherent theoretical and experimental difficulties in the establishment of accurate absolute values of photoelectric cross-sections. In particular it should be noticed that the use of theoretical total cross-sections neglects the change in angle between the direction of the photoelectron and the incident gamma-ray with the energy of the gamma-ray. For practical arrangements this becomes very important at low energies (8).

The conventional method of estimating photo-line intensities is to evaluate the areas under the peaks. It proved, however, easier to use the peak heights as determined from the counting rates at the line peaks and at the constant backgrounds on their high energy sides. In this way, distorting effects on the low energy side do not influence the result. For the calibration lines, Cs^{137} and Co^{60}, the areas could be accurately determined and it was found that these were, in fact, proportional to the product $R\,\Delta H\varrho/H\varrho$, to within 7–8 %. Here R = peak counting rate of the line and $\Delta H\varrho$ = total momentum spread at half maximum, i.e. $\Delta H\varrho/H\varrho$ is the conventional half width of the line. In order to reduce individual errors as far as possible, all half widths were plotted versus $H\varrho$, and a curve was drawn through the actual region to give mean values. This is shown in Fig. 4.

Although the points scatter the general trend is quite obvious: $\Delta H\varrho/H\varrho$ decreases somewhat as $H\varrho$ increases. This is to be expected on account of smaller amounts of scattering and energy loss at higher energies, and so these phenomena are obviously reflected in the curve of Fig. 4. By taking values of $\Delta H\varrho/H\varrho$ from this curve a product $R\cdot\Delta H\varrho/H\varrho$ was assigned to each photo-line in order to represent quantities approximately proportional to the line areas.

The ratios I_m/I_n of the absolute gamma-ray intensities were found by dose rate measurements (14) performed with an ionization chamber.[1] This chamber was known to be energy independent in the region of interest, i.e. it was very nearly air equivalent (the röntgen unit is defined with respect to ionization in air). The chamber

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[1] This chamber was constructed by Dr. N. H. Moxnæs, Statens Fysiska Kontrollaboratorium, Oslo, Norway.

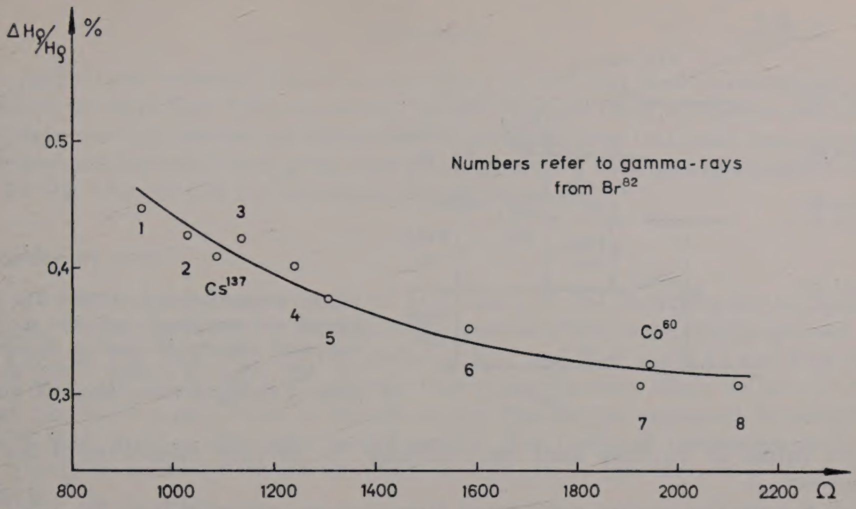

Fig. 4. $\Delta H\varrho/H\varrho$ at half maximum as a function of $H\varrho$ for the observed conversion lines (the abscissa is very nearly proportional to $H\varrho$ as can be seen from Fig. 3).

was calibrated with standard radium sources and had a sensitive volume of 600 cm^3 and a capacitance of about 50 $\mu\mu$F. The comparison of the absolute intensities of the standard Cs^{137} and Co^{60} sources was made from several determinations. Care was taken to minimize the amount of surrounding scattering material and, by trying different source-chamber distances, it was found that no saturation effects of the chamber were present. Furthermore, to avoid possible errors in the calibration of the charging unit, irradiation times were selected so as to result in nearly the same discharge of the chamber. In this way the fiber of the charging unit was always observed in a very small region of the total scale.

Knowing the relative intensities of two gamma-rays and of their external conversion photo-lines, Eq. (3) was used to evaluate relative photoelectric cross-sections. For the reasons mentioned earlier, this was done for the Cs^{137} and the Co^{60} calibration energies. It was then found that the experimental value of the ratio of Eq. (3) for U differed by about 13 % from the theoretical figure (reference (13), p. 870). This deviation is probably due to the above-mentioned angular dependence of the photoelectric cross-section upon the gamma-ray energy and thus our empirical

Table 2. Relative intensities of the gamma-rays from the decay of Br^{82}.

Gamma-ray	Relative intensity
1	80
2	50
3	33
4	100
5	30
6	35
7	32
8	21

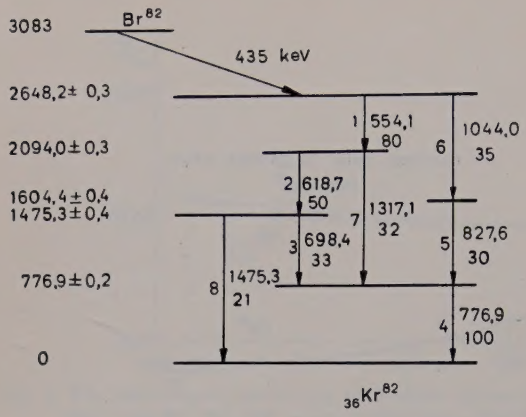

Fig. 5. Decay scheme of Br^{82}. The upper figures are energies in keV and the lower represent relative intensities. Figures on the left refer to Tables 1 and 2. The β-decay energy is taken from ref. (15).

relative values of τ_K were used for evaluting the relative intensities of the Br^{82} gamma-rays.

Table 2 gives the relative gamma-ray intensities as computed from Eq. (2).

Decay scheme of Br^{82}

Our measurements on Br^{82} are in good agreement with the decay scheme shown in Fig. 5.[1] It should be mentioned that our experiments permit no conclusions about the relative positions of the gamma-rays 5 and 6 to be drawn. The fact that the beta-spectrum of Br^{82} is simple (1, 15) and that the internal conversion of the gamma-rays is negligible permits the use of our relative intensity determinations for checking the decay scheme directly.

The internal conversion of the gamma-rays has been roughly estimated on the assumption that, according to the shell model, the first excited state of $_{36}Kr^{82}_{46}$ has the assignment 2+ (valid in more than 95 % of all known first excited states of even-even nuclei (16)) and is expected to be neglibibly low. Thus, the gamma-ray intensities of Table 2 will give the correct transition probabilities within the experimental accuracy. The intensity ratios are expected to be correct to $\sim 10\,\%$.

A calculation of the theoretical dosage rate K_s in röntgens per hour × mC at 1 cm distance from a point source (17) was made for Br^{82}. Using energy and intensity values from Tables 1 and 2 above, we obtain

$$K_s = 14.4 \pm 0.1 \text{ r/(hour} \cdot \text{mC) at 1 cm.}$$

Using a 4π GM counter and $\beta\gamma$-coincidences for the absolute determination of source activity, W. E. Perry (17) obtained the experimental value

$$K_s = 14.7 \pm 0.2 \text{ r/(hour} \cdot \text{mC) at 1 cm.}$$

The agreement is thus good.

Our gamma-ray energies and the most recent investigation of the beta-decay of Br^{82} (15) give the energy difference between the ground states of Br^{82} and Kr^{82} as 3083 keV, according to Fig. 5. This results in the following mass for Br^{82} from that of Kr^{82} (18):

$$\text{Mass } Br^{82} = 81.93967 + 0.00332 = 81.94299 \text{ amu.}$$

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[1] Fig. 5 was obtained by inverting the decay scheme of D. C. Lu *et al.* (see Note added in proof).

Summary

The gamma-ray emission following the decay of Br^{82} has been investigated. The energies of the rays have been accurately determined and their intensities estimated. A calculation of the theoretical dosage rate in röntgens/(hour · mC) at 1 cm in air from our energy and intensity data gives very good agreement with an experimental value from 4π GM counter and $\beta\gamma$-coincidence measurements.

Note added in proof

The inversion of the decay scheme of D. C. Lu *et al.* (6), resulting in the diagram of Fig. 5, was undertaken for the following reasons which were kindly pointed out to us by C. S. Wu. The first excited states of Kr^{80} and Kr^{84} are 0.62 and 0.89 Mev, respectively, and thus the first excited state of Kr^{82} is very likely to be 0.78 MeV so that the three points lie on a smooth curve. The second argument for assigning the 0.78 MeV level as the first excited state is that Coulomb excitation on Kr by alpha particles failed to reveal any line of 0.55 MeV, but there is a line around 0.78 MeV.

A very recent investigation of Br^{82} has been made by R. C. Waddell and E. N. Jensen. Phys. Rev. *102*, 816 (1956). They report internal conversion measurements made in an intermediate image spectrometer (resolution set to 3 %), coincidence measurements and angular correlations between gamma-rays. The gamma-ray intensities were determined from a scintillation spectrum. They found a simple beta-ray spectrum with an end-point energy of 444 ± 1 keV. These authors also point out reasons for inverting the decay scheme of D. C. Lu *et al.* and they give spin and parity assignments for the observed levels in Kr^{82}.

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